Seasonal Variations and Flux of Arsenic in Gomati River, Ganga Alluvial Plain, Northern India

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Introduction

Arsenic is a naturally occurring toxic metalloid and its contamination in the hydrological system has significantly received worldwide attention in the last three decades. It is widely distributed in all the components of the environment and varies by more than four orders of magnitude ranging from <0.5 μg/l to 5000 μg/l in the natural water systems. Natural systems, with climate as the controlling mechanism, play an important role in strong geochemical fractionation with quantitative elemental transportation (Raju, 2012a). Natural systems of tropical environment are, therefore, a major concern for environmental scientists (Dissanayake and Chandrajith, 1999; Smedley and Kinniburgh, 2002). The study of fluvial time series provides the key for the understanding of elemental mobilization that controls the dissolved elemental concentration in the various components of the hydrological system (Raju 2012a). Studies have shown that dissolved As concentration in river water varies at a great extent and is mainly dependent on geology, hydrology, climate as well as various anthropogenic activities (Raju 2012b; Masson et al., 2007; Elbaz-Poulichet et al., 2006; Pettine et al., 1997; McLaren and Kim, 1995). In northern India, the Ganga Alluvial Plain (GAP) is one of the most densely populated regions of the world. It is drained by several alluvial rivers and supports nearly 500 million people. The objective of this paper is to report

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the seasonal variation and flux of dissolved As in the Gomati river and to understand the As mobilization for its eco-toxicological potentials in the GAP.

Study Area

The Gomati river (an alluvial river of the GAP) originates from the Gomath Tal and covers a distance of 900 km before its confluence with the Ganga river. It drains about 30,437 km^2 area, located between 25–29°N and 80–84°E in the interfluve region of the Ganga and Ghaghara rivers and encompasses an altitude range of 190 m to 60 m above mean sea level (Fig. 1). Geologically, the Gomati River Basin is made up of unconsolidated alluvial sediments derived from the Himalayan region. These alluvial sediments are made up of well sorted silt and silty fine sand, representing muddy and sandy interfluve deposits (Singh, 1996). Silt fraction of these alluvial sediments is predominantly composed of mica along with feldspar, quartz and mixed layer minerals in the GAP (Pal et al., 2011).

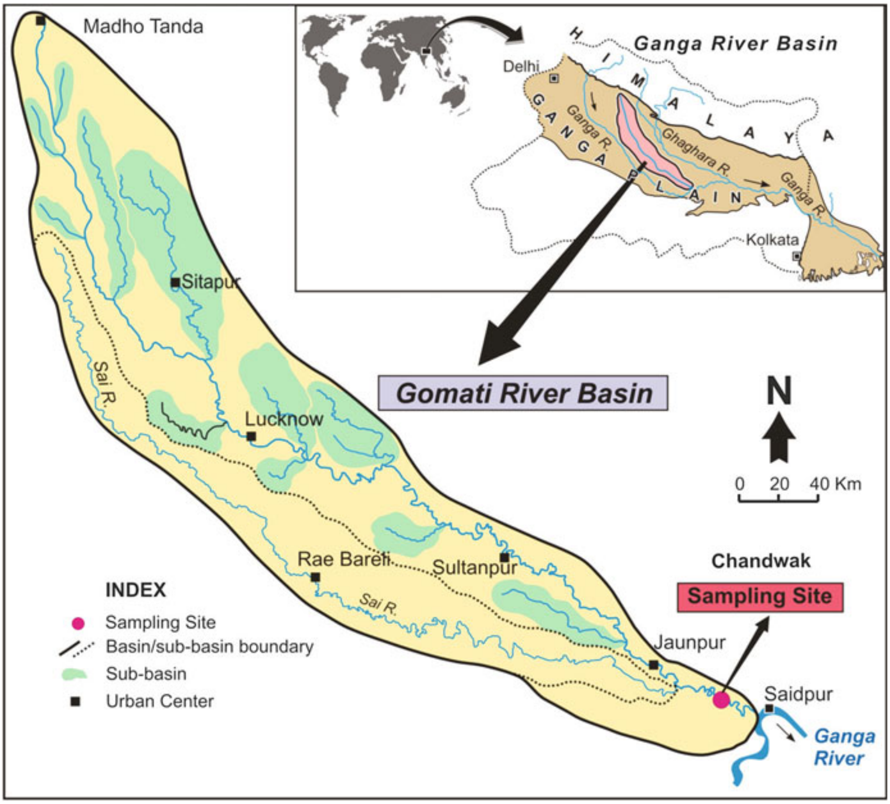

Fig. 1 Location map of the study area. The Gomati river drains the interfluve region of the Ganga and the Ghaghara rivers in the Ganga Alluvial Plain, northern India. Sampling site Chandwak was selected for the present study

The Gomati river basin experiences a warm and humid sub-tropical climate with four prominent seasons: the winter, the summer, the monsoon and the post-monsoon seasons (Dasgupta, 1984). The winter season is a cold dry season that extends from December to February, characterized by low temperature and low rainfall. The hot dry summer season extends from March to May and is marked by a continuous rise in temperature that reaches a maximum of 40 to 47 °C in May. The monsoon season is a warm wet season, extending from June to September. It is characterized by high rainfall due to the southwest monsoon system. Daily mean maximum temperature is about 30 °C. Nearly 70 – 80 % of the total annual rainfall occurs during this season. The Gomati river along with its tributaries and other water bodies including the low-lying areas in the regional surface of the GRB get flooded due to excessive monsoon rainfall. The post-monsoon season extends from October to November and is characterized by the decreasing trend of maximum (35 – 25 °C) and minimum (20 – 10 °C) temperatures along with moderate rainfall.

The Gomati river is a groundwater fed river. Its hydrology is strongly controlled by the seasons. The river discharge varies from 31.85 m^3/s in the summer season, to 721.32 m^3/s in the monsoon season. The first monsoon peak discharge (>600 m^3/s) occurs in the end of July and is ~20 times higher than the average discharge (30 m^3/s) during the summer season (Table 1). The $\delta^{18}O$ characterization of precipitation recorded at New Delhi shows the lower-most values in September (late monsoon) and November (post-monsoon) months (IAEA, 1981). Low isotopic characters ($\delta^{18}O = -10$ ‰) of the monsoon precipitation pass into the shallow groundwater and the Gomati river water as the monsoon precipitation is the dominant process of the recharge activity of the Gomati river basin in the GAP.

Methodology

Chandwak (25°35′ N, 83°00′ E) has been selected to represent the entire drainage basin and is located upstream of the confluence of the Gomati with the Ganga river at an elevation of 70 m above mean sea level (Fig. 1). Thirty-six river water samples were collected from the middle of the river channel during June 2009-May 2010 at an interval of ten days. The river water samples were collected in wide mouth 250 ml polypropylene bottles (© Tarsons) with airtight caps and were acidified in the field with HNO_3 (5 ml/l). Each sample bottle was tagged with appropriate label and details. All these water samples were carefully transported into the laboratory and stored at 4 °C in the laboratory till further chemical analysis. The water samples were filtered by using 0.45 μm cellulose filters. All the filtered samples were analysed by Induced Coupled Plasma-Mass Spectrophotometer at the Institute Instrumentation Centre, Indian Institute of Technology, Roorkee. Each sample was analysed in duplicate and mean values were taken as the result. All the samples were analysed in the laboratory following the standard protocols (APHA, 2002). For quality assurance, replicates and analytical blanks were also prepared and analysed to check the

Table 1 Sampling details and dissolved As concentration (μg/l) along with discharge and flux of the Gomati river at Chandwak

Sample code	Sampling date (dd.mm.yy)	Season	Dissolved As (μg/l)	Discharge (m^3/s)	Flux (kg/day)
GRW01	05.06.09	Monsoon	4.34	88.17	33.06
GRW02	14.06.09	Monsoon	2.41	95.86	19.96
GRW03	25.06.09	Monsoon	2.43	63.76	13.39
GRW04	05.07.09	Monsoon	3.80	66.03	21.68
GRW05	15.07.09	Monsoon	3.27	95.02	26.85
GRW06	25.07.09	Monsoon	1.306	11.73	68.71
GRW07	05.08.09	Monsoon	1.012	09.91	18.32
GRW08	16.08.09	Monsoon	2.112	27.13	41.41
GRW09	25.08.09	Monsoon	1.26	232.66	25.33
GRW10	05.09.09	Monsoon	2.40	409.92	85.00
GRW11	16.09.09	Monsoon	0.5371	1.83	32.60
GRW12	25.09.09	Monsoon	1.01	380.57	33.21
GRW13	05.10.09	Post-monsoon	1.28	721.32	79.77
GRW14	17.10.09	Post-monsoon	0.98	697.56	59.06
GRW15	25.10.09	Post-monsoon	0.83	280.80	20.14
GRW16	05.11.09	Post-monsoon	1.06	174.49	15.98
GRW17	15.11.09	Post-monsoon	1.61	91.73	12.76
GRW18	25.11.09	Post-monsoon	1.93	78.59	13.11
GRW19	05.12.09	Winter	1.61	110.45	15.36
GRW20	15.12.09	Winter	1.18	168.70	17.20
GRW21	25.12.09	Winter	1.37	102.34	12.11
GRW22	05.01.10	Winter	1.25	128.50	13.88
GRW23	15.01.10	Winter	1.28	164.37	18.18
GRW24	25.01.10	Winter	1.18	145.30	14.81
GRW25	05.02.10	Winter	1.27	61.42	6.74
GRW26	15.02.10	Winter	1.89	132.70	21.67
GRW27	25.02.10	Winter	1.57	98.49	13.36
GRW28	05.03.10	Summer	2.19	63.95	12.10
GRW29	15.03.10	Summer	2.59	65.11	14.57
GRW30	25.03.10	Summer	2.65	53.30	12.20
GRW31	05.04.10	Summer	3.31	60.17	17.21
GRW32	15.04.10	Summer	3.55	42.67	13.09
GRW33	25.04.10	Summer	4.71	36.59	14.89
GRW34	05.05.10	Summer	5.15	35.84	15.95
GRW35	15.05.10	Summer	5.18	31.85	14.25
GRW36	25.05.10	Summer	5.67	33.50	16.41

Note: The office of Middle Ganga Division, Central Water Commission provided the discharge data for the Gomati river at Maighat, Chandwak

reliability of the data. Analytical precision for all samples were within ±5 % in As determination. Dissolved As concentration and discharge data were used to estimate the dissolved flux by using commonly applied equations (Table 1).

Results

Dissolved As Concentration and Seasonal Variations

The dissolved As concentration in the river water varies from 0.53 to 5.67 μg/l, whereas baseline concentrations of As in the world's river waters were very low, about 0.1-0.8 μg/l (Smedley and Kinniburgh 2002). The dissolved As concentrations (<45 μm) varied by a factor ~6 in the Gomati river (Table 1). Figure 2 displays variations of dissolved As concentration in the Gomati river water during the monsoon, the post-monsoon, the winter and the summer seasons. The variations in dissolved As concentration have been already reported for the Garonne, the Dordogne and the Isle rivers in France and also for the Seine, Po and Waikato rivers (Masson et al., 2007; Elbaz-Poulichet et al., 2006; Pettine et al., 1997; McLaren and Kim, 1995). During the winter season, the dissolved As concentration in the Gomati river water was stable around 1.00 to 2.00 μg/l. In the summer season, As concentration continuously increases from 2.19 to 5.67 μg/l. This is the most significant characteristic of As variation in the Gomati river water which is due to increasing temperature in the summer season. High temperature leads to the enormous growth of microbial species within the river sediments which facilitates the mobilization of As from the river sediments to the Gomati river water (Oremland and Stolz, 2003; Islam et al., 2012).

During the monsoon and the post-monsoon seasons, dissolved As concentration showed cyclic decreasing trend from 4.34 to 0.53 μg/l. It is mainly dependent on the monsoon rainfall patterns and flood discharges that mainly control the hydrology of the river. Dissolved As concentrations, therefore, were affected by dilution in these seasons. These results suggest that the seasonal variations in dissolved As

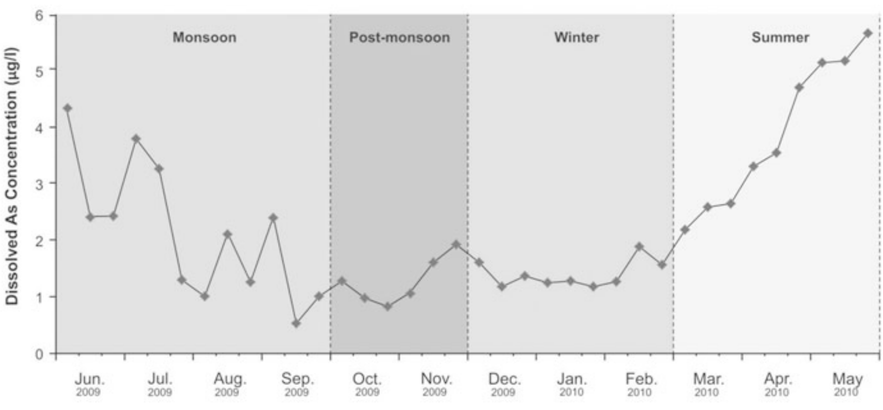

Fig. 2 Seasonal variation of dissolved As concentration (μg/l) in the Gomati river water at Chandwak during June 2009 to May 2010. Note the steady trend during the winter, continuous increasing trend during the summer and cyclic decreasing trend during the monsoon and the post-monsoon seasons

concentration show a single cyclic pattern controlled by the temperature during the winter and summer seasons, and the river's flood discharge during the monsoon and post-monsoon seasons.

Relationship between River Hydrology

The dissolved As concentrations in the Gomati river displayed the moderate negative correlation coefficient ($r^2 = 0.49$) with the river's flood discharges during the monsoon season (Fig. 3a). It can be summarized that dissolved As concentration in the river water truly depends on the river's flood discharge that is controlled by dilution factor through precipitation in the GAP during the monsoon season. During the post-monsoon and winter seasons, insignificant correlation coefficients were recorded (Fig. 3b, c). Significant correlation ($r^2 = 0.86$) has been observed between the river discharge and the dissolved As concentration during the summer season (Fig. 3d). This correlation can be linked with a decrease in the river's discharge along with rise in temperature that controls As mobilization in the river water.

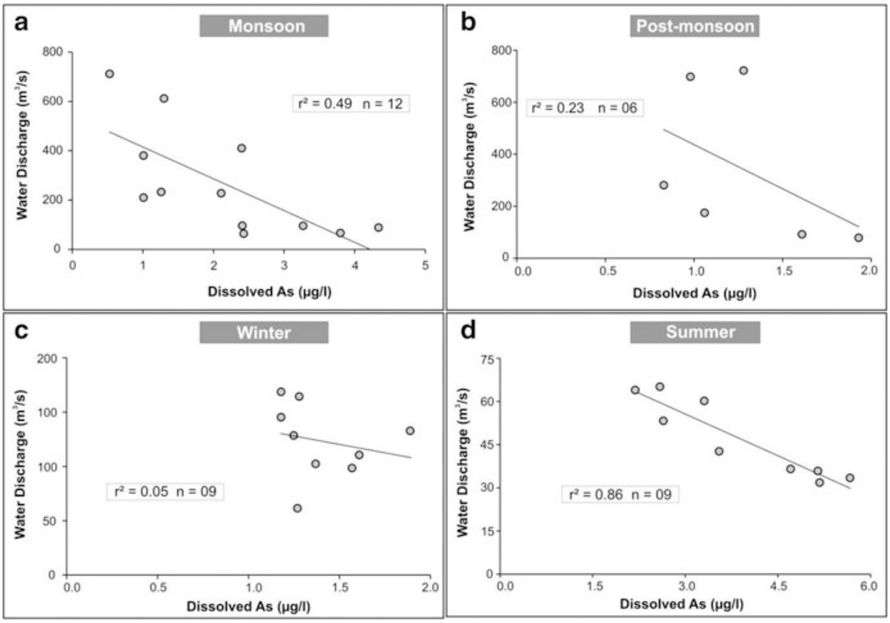

Fig. 3 Correlation plots of water discharge (m^3/s) and dissolved As concentration (μg/l) of the Gomati river in (**a**) the monsoon; (**b**) the post-monsoon; (**c**) the winter and (**d**) the summer seasons

Dissolved as Flux and Its Seasonal Variations

Figure 4a displays a bar diagram representing the monthly variations in the dissolved As flux of the Gomati river. The estimated total dissolved As flux of the Gomati river is 8.99×10^3 kg that is transported into the Ganga river during June, 2009-May, 2010. The fluxes of dissolved As of the Gomati river were recorded to be 4289 kg during the monsoon, 2028 kg during the post-monsoon, 1333 kg during the winter and 1335 kg during the summer seasons. The Gomati river contributes ~70 % of total dissolved As flux during the monsoon and post-monsoon seasons and ~30 % during the winter and summer seasons (Fig. 4b). This is due to the fact that Gomati river basin receives up to 80 % of the annual rainfall from June to September. The As mobilized from the top alluvial sediments of the GAP may be contributed to the dissolved As fluxes of the Gomati river during the monsoon and post-monsoon seasons, particularly after heavy precipitation events. This clearly indicates that the monsoonal rainfall and surface runoff contributes greater amount of As into the Gomati river.

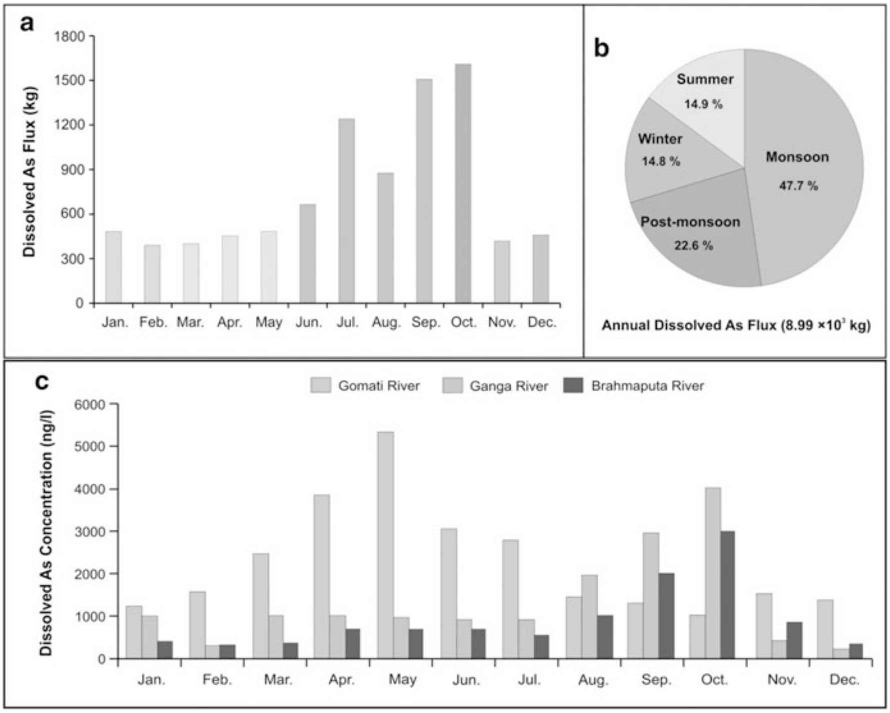

Fig. 4 (**a**) Bar diagram showing the monthly distribution of dissolved As flux of the Gomati river to the Ganga river; (**b**) Pie diagram showing the seasonal distribution of dissolved As flux of the Gomati river. Nearly 70 % of the total annual As flux (8.99×10^3 kg) is transported during the monsoon and post-monsoon seasons. (**c**) Bar diagram representing comparative variations in dissolved As concentration (ng/l) in the Gomati, the Ganga and the Brahmaputra rivers. [Data source: Islam et al., 2012]

Discussions

Comparative Study with the Ganga and Brahmaputra Rivers

The dissolved As concentrations showed seasonal variations ranging from 0.2 to 4.0 μg/l for the Ganga and 0.3 to 3.0 μg/l for the Brahmaputra rivers (Islam et al., 2012). Figure 4c displays the bar diagram showing monthly variations of dissolved As concentrations in the Gomati along with the Ganga and the Brahmaputra rivers. Except during the monsoon season, dissolved As concentrations in the Gomati river water is reported to be higher than the Ganga and the Brahmaputra rivers and are linked with the characteristics of the river hydrology and anthropogenic influences.

Empirical Modelling

Figure 5 displays an empirical model to explicate the seasonal variations of dissolved As concentration and flux of river in the GAP, northern India. The geogenic source of As in the GAP is most likely alluvial sediments forming mineral mica (muscovite and biotite), derived from the granitic and metamorphic source regions of the Himalaya (Breit, 2000). Average As concentrations in the GAP sediments was estimated to be about 10.44 mg/kg (Singh et al., 2012). The Gomati river bed sediments contain 1.36 mg/kg and the Gomati river suspended sediments 5.30 mg/kg (Singh et al., 2012). In the Gomati river water, dissolved As is released from unconsolidated alluvial sediments of the GAP through natural chemical weathering process. Mineral biotite is considered as source of As in the unconsolidated sediments of the GAP (Seddique et al., 2008; Chakraborty et al., 2011; Singh et al., 2013). Biotite weathers very easily and may be converted to kaolinite. In the process of kaolinization of biotite, the weathering reaction requires chemical changes by addition of H^+ and release of K^+, Al^{3+}, Mg^{2+}, Fe^{2+}(As), Ti^{4+}, Si^{4+} and H_2O (Dong et al., 1998).

$$
\begin{aligned}
&K_{0.65}(Al_{1.10}Ti_{0.15}Fe_{0.50}\,Mg_{0.55})(Si_{3.20}Al_{0.80})O_{10}(OH)_2 + 8H^+ \\
&\quad = (Al_{1.71}Fe_{0.31}Ti_{0.03})Si_2O_5(OH)_4 + 0.65\,K^+ + 0.19Al^{3+} + 0.55\,Mg^{2+} \\
&\qquad + 0.19Fe^{2+} + 0.12Ti^{4+} + 1.2Si^{4+} + 3H_2O
\end{aligned}
$$

Reductive dissolution is the established geochemical mechanism that mobilizes As from the Gomati river and alluvial sediments into the Gomati river water. It occurs in the presence of Fe containing silicate minerals, which break down under the influence of decaying organic matter. This chemical process is driven by the decay of organic matter (Ravenscroft et al., 2009). Microbes (such as cyanobacteria, algae and lichens etc.) are essentially ubiquitous in sediments and inhabit on the surface of minerals. These microbes dissolve silicate minerals through the corrosive action of metabolic products, such as NH_3, HNO_3 and CO_2

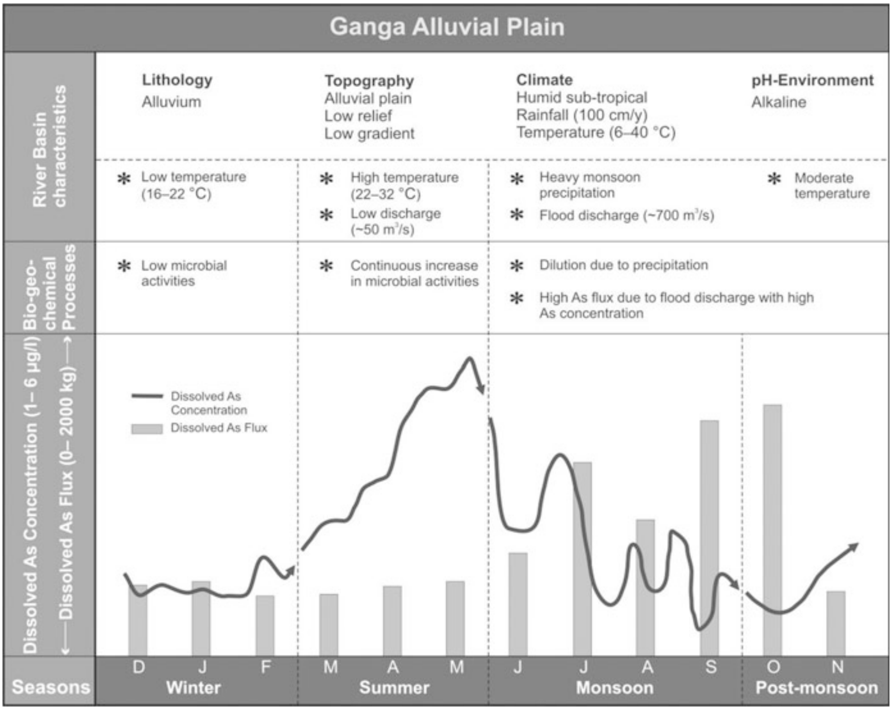

Fig. 5 Empirical model showing the characteristics and biogeochemical processes involved in the seasonal variation of dissolved As concentration and flux of river in the Ganga Alluvial Plain, northern India

forming H_2CO_3 in water etc. These metabolic products, execrated by micro-organisms that promote the chemical weathering of primary minerals, result in the displacement of cationic components from crystal lattice due to breaking of Si-O and Al-O bonds. Microorganisms, therefore, directly or indirectly, induce mineral disaggregation, hydration, dissolution and secondary mineral formation (Banfield et al., 1999; Ehrlich and Newman, 2009). The Gomati River receives organic matter rich untreated urban effluents from major urban centres located on its banks such as Lucknow, Sultanpur, Jaunpur etc. Singh et al. (2010) recently reported the downstream increase in the dissolved As concentration from 1.3 to 9.6 µg/l in the Gomati river during the winter season. The microbiological activity, therefore, plays a significant role in the mobilization of As from the sediments and dissolved As concentrations in the river water during the winter and summer seasons.

During the monsoon season, rainwater becomes either groundwater or floodwater after interacting with mica dominated silty alluvial sediments of the GAP after the 3-month long summer season. Robert et al. (2011) recently reported that monsoon derived floodwaters in the Ganga Delta region recorded high As concentrations (~500 μg/l). The recent $\delta^{18}O$ isotopic study of the Gomati river basin displayed that there is an instantaneous exchange between shallow groundwater and river water through precipitation during the monsoon season (Singh et al., 2013). The duration and intensity of precipitation is accountable for the river hydrology and also governs the dilution of dissolved As concentration due to the rapid exchange of the monsoon precipitation into the river water through the shallow groundwater. It can also be justified through similar oxygen isotopic compositions of the shallow groundwater and the Gomati river water (Singh et al., 2013). From the above studies, it can be concluded that heavy monsoon precipitation, instantaneous interactions of shallow groundwater and the Gomati river water, high As concentrations in flood waters are responsible for the significant dissolved As flux of the Gomati river during the monsoon and post-monsoon seasons.

Conclusions

The present study reveals that dissolved As in rivers of the GAP show significant seasonal variations controlled by the range and duration of temperature and monsoon precipitation. Nevertheless, all dissolved As concentrations in the Gomati river were higher than the Ganga and the Brahmaputra Rivers during the summer and winter seasons that can be linked with hydrological and biogeochemical factors. The Gomati River substantially contributes the dissolved As flux to the Ganga river. Anthropogenically induced organic matter rich untreated effluents draining into rivers of the GAP govern the fate and mobility of As in the fluvial environment of the GAP during the summer season. Our findings may have important implications for the designing of research and monitoring programmes in As-affected areas of the GAP as well as other sub-tropical regions of the world.

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